Dust Condensation of SiC, SiO in Asymptotic Giant Branch Stellar Winds - SiC Spectrum Postprint

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Abstract

We have chosen the Large Scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) code to calculate the coalescence of silicon carbide (SiC), silicon oxide dust (SiO) in the AGB stellar wind. LAMMPS is a classical molecular dynamics simulation code. At the same time, we consider the effect of temperature on the evolution of molecular dynamics. We also calculated the temperature change of non-spherical SiC, SiO dust coalescence. The condensation temperature range of SiC dust in the AGB stellar wind is 300–500 k and 900–1100 k for SiO. Finally, the infrared spectrum of SiC was calculated using Gaussian 16 software. The 77SiC, 70Si3C3, and 121Si3C3 models have clear characteristic peaks of infrared spectra responding at 5, 8.6, 11.3, 15, 19, and 37 m.

Full Text

Preamble

Dust Condensation of SiC, SiO in Asymptotic Giant Branch Stellar Winds—SiC Spectrum

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Abstract

We employ the Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) code to investigate the condensation of silicon carbide (SiC) and silicon monoxide (SiO) dust in asymptotic giant branch (AGB) stellar winds. LAMMPS is a classical molecular dynamics simulation code that enables us to examine temperature effects on the dynamical evolution of these systems. We calculate the condensation temperature ranges for SiC and SiO dust in AGB stellar winds, finding [300–500] K for SiC and [900–1100] K for SiO. Finally, we compute the infrared spectra of SiC using Gaussian 16 software. The 77SiC, $70Si_3C_3$, and $121Si_3C_3$ models exhibit clear characteristic infrared spectral peaks at 5, 8.6, 11.3, 15, 19, and 37 m.

Key words: stars: AGB and post-AGB – infrared: ISM – (ISM:) dust – extinction

1. Introduction

During the asymptotic giant branch (AGB) phase, stars with initial masses up to 8 M lose their outer layers through stellar outflows or winds, creating extended circumstellar envelopes (CSEs). These CSEs serve as rich astrochemical laboratories where over 100 molecules and newly formed dust species have been detected. The chemical composition of a CSE is determined by the elemental carbon-to-oxygen ratio (C/O) of the AGB star itself, with $C/O < 1$ leading to oxygen-rich outflows and $C/O > 1$ leading to carbon-rich outflows (see, e.g., Habing & Olofsson 2003).

During the thermally pulsing AGB phase, low- and intermediate-mass stars develop strong mass loss and eject their envelopes. These stars become enshrouded by optically thick dust shells, where solids condense from the gas phase as tiny dust grains (Bowen 1988; Gail & Sedlmayr 1999). Dust is broadly classified into O-rich and C-rich varieties. The pure thermal decomposition of olivine results in the formation of silicon monoxide dust (SiO), along with Mg, Fe, and O (Gail & Sedlmayr 1999). C-rich dust consists primarily of amorphous carbon and silicon carbide (SiC; Van de Sande et al. 2019).

Since no condensation seeds exist in the gas simply outflowing from the star, silicate dust grains eventually grow through a series of physicochemical events in cooler outflows. The first step in this sequence must be the formation of seed particles. Because SiO is the most abundant gas-phase refractory element that may participate in this process, considerable discussion has focused on whether clusters formed from SiO serve as the basis for all subsequent dust formation processes (Gail & Sedlmayr 1986; Ali & Castleman 2005; Nuth & Ferguson 2006; Paquette et al. 2011; Goumans & Bromley 2012). Gail et al. (2013) established an upper limit to SiO stability based on new measurements of the vapor pressure of SiO molecules against solid SiO. SiO may indeed be the first substance to nucleate out of a mixture of oxygen-rich elements (Nuth & Donn 1981).

The dominant population (>0.9) of presolar silicon carbide (SiC) is believed to originate from low-mass AGB stars with near-solar metallicities (Liu & Stephan 2022). Nearly a century ago, Wildt (1933) considered the possibility that solid SiC grains might form in N-type stars. It is now recognized that SiC solids constitute a major dust species—second only to amorphous carbon particles—condensing in the cold atmospheres of mass-losing, carbon-rich AGB stars (Nanni et al. 2021; Chen et al. 2022). Spectroscopic observations have revealed the widespread presence of SiC dust in carbon stars, characterized by a prominent 11.3 m emission feature attributed to Si–C stretching modes in SiC solids (Speck et al. 1997; Mutschke et al. 1999). However, the exact mass fraction of SiC condensate in carbon stars remains uncertain and depends on stellar mass and metallicity (Nanni et al. 2021). Radiative transfer modeling of observed infrared emission from carbon stars suggests that the mass ratio of SiC to amorphous carbon may be as high as 0.25 in the Milky Way (Groenewegen et al. 1998), 0.43 in the Large Magellanic Cloud, and 0.11 in the Small Magellanic Cloud (Groenewegen et al. 2009; Nanni et al. 2019). Theoretical dust yield calculations predict that SiC comprises 0.25 of the total dust mass (silicon carbide plus amorphous carbon) for carbon stars with an initial mass of 3 M at solar metallicity (Nanni et al. 2013; Zhukovska & Henning 2013).

Small molecules containing silicon and carbon are thought to be important components of interstellar particles. Some have been detected in the circumstellar environments of evolved stars through rotational spectroscopy, such as SiC, SiC_2, Si_2C, c-SiC_3, and SiC_4, while centrosymmetric species such as C_3, C_4, C_5, Si_2C_2, and Si_2C_3 can only be detected through vibrational transitions (primarily in the infrared) (Witsch et al. 2017). Witsch et al. (2017) presented laboratory data on the asymmetric stretching mode of Si_2C at 8.6 m, using a pulsed Nd:YAG laser to vaporize a solid Si target in a dilute methane sample exposed to helium buffer gas. Si_2C was formed in the adiabatic expansion of a supersonic jet, and rotationally resolved spectra were recorded using radiation from a quantum cascade laser.

Theoretical calculations (Gail & Sedlmayr 1986) based on the results of Nuth & Donn (1981) suggest that SiO nucleation should not begin until temperatures drop below 600 K, whereas observed condensation temperatures of periastron dust shells are typically in the range of 900 K and sometimes even higher. The total ice number density and number of monolayers throughout the outflow depend on the dust temperature profile, with clear differences between amorphous carbonaceous grains and SiC dust (Van de Sande et al. 2019). According to Van de Sande et al. (2019), the dust temperature profile for C-rich dust (SiC) is 350–

500 K, while for O-rich dust it is 550–800 K. The C-rich dust primarily consists of large quantities of amorphous carbon and SiC.

The fundamental process of primary grain condensation can be best illustrated by considering stellar winds, where both gas temperature and material dilution phenomena occur. There is a monotonic decrease in both gas temperature and matter density with increasing distance from the star, leading to recombination of the plasma and the sequential appearance of diatomic, polyatomic, and larger molecules. At certain distances where temperatures fall well below 1500 K, atoms and molecules combine into stable clusters, which can then grow into homogeneous or heterogeneous macroscopic samples through the addition of further similar or dissimilar substances (Sedlmayr 1994).

In recent years, numerous research groups have investigated the dust and heavy element yields (^{1}H, ^{12}C, ^{13}C, ^{14}N, ^{15}N, and ^{16}O) from AGB stars (Ferrarotti & Gail 2006; Karakas & Lattanzio 2007; Ventura et al. 2020, 2021; Cinquegrana & Karakas 2022) and supernovae (Wu et al. 2021). We previously estimated the abundance of SiC in the solar system based on spectral analysis, provided that the spectrum was properly corrected.

Until now, only a few groups have studied the SiC and SiO dust condensation processes in AGB stellar winds and the infrared spectra of SiC dust. Investigating the role of temperature in molecular dynamics for both SiC and SiO, along with comparative analysis of their resulting radiation spectra, holds significant research importance. Understanding the content, structure, and spectra of SiC has profound implications for astrophysics. The topic of SiC and SiO dust condensation is particularly important and fascinating because it enhances our understanding of the origin of these species, the fundamental laws governing stellar evolution, and the evolution of the interstellar medium. Such studies can inform observations with the James Webb Space Telescope (JWST) regarding dust structure and composition.

2. Models and Methods

We employ the Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) code to compute the SiC and SiO dust condensation processes in AGB stellar winds. LAMMPS is a classical molecular dynamics (MD) simulation code designed for efficient execution on parallel computers, with capabilities for easy extension and modification. It can calculate various material properties including viscosity, diffusion coefficients, energy, radial distribution functions, and the melting and solidification behavior of spherical or ellipsoidal particles (Plimpton 1995; Denniston & Navid 2022).

The simulation parameters are configured as follows: SiC and SiO atoms are uniformly distributed with an initial spacing of 2 Å in a simulation box of size 20 × 20 × 20 Å^{3} with periodic boundaries. The particle heat bath box size is 40

$\times$ 40 $\times$ 40 Å^3. The timestep is 0.05 fs, and atomic velocities follow a Gaussian distribution. We employ the NPT ensemble to investigate condensation of SiC and SiO dust at various temperatures and pressures. The condensation temperature ranges are [300–500] K for SiC dust and [900–1100] K for SiO dust in AGB stellar winds. The SiC and SiO dust models contain 100 and 121 atoms, respectively.

For the interatomic potential, we utilize the ReaxFF force field, which has been employed by David et al. (2012) to study the initial oxidation of SiC surfaces by O_2 and H_2O molecules. ReaxFF naturally simulates bond breaking and formation, and can describe Si–C–O and Si–O–H bond interactions. Based on the principle of determining bond orders according to interatomic distance, ReaxFF describes chemical system interactions smoothly across bonding transitions. Compared with conventional force fields, ReaxFF provides better description of polarization because atomic charges are dynamically optimized using the electronegativity equalization method at each simulation timestep. Relative to quantum mechanics, ReaxFF molecular simulation is more suitable for complex systems because reaction paths are determined entirely by system dynamics (Du & Wang 2018).

LAMMPS numerically integrates Newton's equations of motion for particles such as atoms, molecules, or macroscopic particles. At the beginning of each simulation, expressions for the resolved forces and potential energies at discrete coordinate positions are computed and stored in arrays. During the simulation, spline interpolation is used to obtain accurate force and potential energy values.

The mean square displacement (MSD) characterizes the average squared distance traveled by particles over time interval t. The diffusion coefficient is given by $\langle r^2(t)\rangle = 2dD$, where d is the dimensionality. The velocity autocorrelation function (VACF) is defined as $\langle v(t)v(0)\rangle$.

Infrared radiation spectra are obtained through Gaussian 16 calculations. Gaussian software applies uniform broadening using a Lorentzian line shape to simulate realistic spectral lines. Generally, electronic spectra (UV-Vis, ECD) are broadened with Gaussian functions, while vibrational spectra (IR, Raman, VCD) are broadened with Lorentzian functions, which decay more slowly than Gaussian functions. According to the uniform bandwidth Lorentzian line equation, the wavenumber is $v = 5.3/\tau$ (cm^{-1} ps^{-1}) for a quantum state with lifetime τ. When the frequency mean interval w^2 is taken as 10, and using half the radiation intensity $I(\omega)/2$, we can derive the bandwidth. For $I(\omega)$, the bandwidth range obtained from the maximum and minimum radiation intensity values is 0.1–0.5 nm.

Infrared spectral (IR) calculations are performed on the final LAMMPS structures after temperature equilibration. IR spectroscopy probes vibrational transitions by measuring light absorption across a range of infrared frequencies. The total absorbed energy, called the intensity, is plotted as peak height versus frequency, yielding a unique spectral fingerprint of the molecule's constituent atoms and bonding structure. We apply density functional theory (DFT) with the B3LYP functional and the 6–21g basis set. B3LYP is a popular and well-regarded functional particularly suitable for metal oxides and other inorganic compounds, and has been used by other researchers for asymmetric model calculations (Lee et al. 1988). The initial calculation obtains self-consistent energy, optimized atomic configuration, and spin density, requiring convergence of the root-mean-square density matrix to less than 10^{-8}. A single scaling factor of 0.965 is applied to compare DFT harmonic wavenumbers NDFT (cm^{-1}) with experimental data (Ota 2015). The corrected wavenumber N is obtained via N (cm^{-1}) = NDFT (cm^{-1}) × 0.965, and the wavelength λ is calculated as λ (m) = 10,000/N (cm^{-1}) (Ota 2019).

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3. Results

Our MD simulations demonstrate that 100-atom systems of SiC and SiO stabilize into final equilibrium structures. The 77SiC, 70Si_3C_3, and 121Si_3C_3 models exhibit characteristic infrared spectral peaks at 5, 8.6, 11.3, 15, 19, and 37 m, represented by curves of different colors.

3.1. SiC and SiO Molecular Dynamics and Spectral Calculations

The classical dust formation process occurs in two stages: first, critical clusters form as condensation seeds, and second, dust grains grow to macroscopic size through the addition of monomers (Siess et al. 2022). In Figure 1 [Figure 1: see original paper], the initial configuration of the 121Si_3C_3 model is adapted from Wu et al. (2022). We selected Si_3C_3 because it represents the smallest cluster with a three-dimensional structure (Gobrecht et al. 2017), enabling investigation of low-mass cluster spectra. Our rationale for choosing the 77SiC, 70Si_3C_3, and 121Si_3C_3 structures for IR spectral calculations rests on two main considerations: first, few researchers have calculated the IR spectra of these specific molecular structures; and second, these molecules facilitate calculation of non-spherical spectra and allow straightforward replication for established studies.

For LAMMPS simulations, the fundamental principle of molecular dynamics involves using Newtonian mechanical equations to describe atomic positions, velocities, and interatomic interactions. By solving these equations, we can simulate molecular kinetic behavior and compute properties of large-scale systems. Our simulations show that atoms coalesce into small SiC and SiO clusters through intermolecular attractive and repulsive forces. In the gaseous state, smaller intermolecular distances produce stronger interactions. Conversely, when molecules experience high temperature or pressure, intermolecular spacing gradually decreases, generating significant heat exchange. Gravitational forces arise from charge interactions between molecules, and when intermolecular distances stabilize, morphological changes lead to solid formation.

In Figure 2 [Figure 2: see original paper], temperature is calculated by dividing kinetic energy by the number of degrees of freedom (and the Boltzmann constant). Since kinetic energy depends on particle velocity, we distinguish between advective velocity (due to bulk particle motion) and thermal velocity, with the latter representing the temperature of interest. We perform constant NPT simulations to update the position, velocity, and angular velocity of finite-size particles. The initial temperature of SiC molecules decreases from 500 K to 300 K, with pressure effects being negligible (following Hanine et al. 2020). The maximum temperature in the figure can reach 800 K due to strong intermolecular forces that cause temperature increases. Similarly, SiO molecules cool from 1100 K to 900 K, with maximum temperatures reaching 1600 K.

Figure 3 [Figure 3: see original paper] demonstrates that the total energy of both molecular systems reaches stable values, indicating that molecular dynamics has achieved equilibrium. Both models exhibit downward trends toward equilibrium. For example, the final total energy is as low as -1.2×10^4 kcal mol^{-1}. The SiC cluster mass ranges from 3.31×10^{-24} kg, while SiO cluster masses range from 3.65×10^{-25} to 7.3×10^{-25} kg. Total energy includes kinetic energy $E_1 = 0.5mv^2$, potential energy $E_2 = mgh$ (where m is atomic mass, v is velocity, and h is atomic displacement), and rotational energy $E_3 = 0.5I\omega^2$ (where I is the rotational moment of inertia and ω is angular velocity).

In Figure 4 [Figure 4: see original paper], the horizontal axis indicates wavelength and the vertical axis shows radiation intensity magnitude. The 77SiC model exhibits peaks at 6, 8.6, 10, 15, 20, 31, and 33 μm, with a particularly strong characteristic peak at 15 μm attributed to extensive Si–C bond stretching. As shown in Figure 5 [Figure 5: see original paper] (right panel), the Si–C and Si–Si bond length scaling on the right side of the blue solid line is more significant than that on the left side.

The 121Si_3C_3 model shows peaks at 8.6, 12, 16, 19, 23, 27, 29, 37, and 66 μm, with two strong characteristic peaks at 19 and 37 μm due to extensive Si–C and Si–Si bond stretching. As shown in Figure 5 [Figure 5: see original paper] (left panel), the Si–C and C=C bond length scaling on the right side of the blue solid line is more significant than that on the left side.

The 70Si_3C_3 model displays peaks at 5, 11.3, 14, 17, 18, 21, 29, 53, and 60 μm, with two strong characteristic peaks at 5 and 11.3 μm resulting from substantial Si–C bond stretching. Among the three models, the 70Si_3C_3 model shows the highest radiation intensity, followed by the 121Si_3C_3 and 77SiC models in decreasing order.

Regarding the radiative signature of SiC at 11.3 μm (Speck et al. 1997; Mutschke et al. 1999), only our 70Si_3C_3 model exhibits a significant radiative signature at this wavelength; the other two models show only weak features, which we attribute to weaker Si–C bond scaling and stronger Si–Si and C=C bond scaling. Witsch et al. (2017) presented laboratory results for the asymmetric stretching mode of Si_2C at 8.6 μm, and we obtain peaks at this wavelength for both the

77SiC and $121Si_3C_3$ models. These results provide theoretical support for future observations of dust in the interstellar medium and are significant for predicting stellar dust yields, galactic dust budgets, and the influence of dust radiation spectra on observations. They can serve as references for dust observations with JWST, the Large Sky Area Multi-Object Fiber Spectroscopy Telescope (LAMOST), and the upcoming China Space Station Optical Survey Telescope (CSST).

We estimate the abundance of SiC in the solar system based on spectral analysis. The infrared emissivity per unit mass of SiC molecules, mass absorption coefficient, and absorption cross-section can be combined to yield:

If SiC has a radiation flux $F\lambda$ at 11.3 m and the distance between a star and Earth is d, then according to the relevant formulas we can derive:

We correct $F\lambda$ by multiplying it by 10^{-30} to match observed spectra. Using the Earth-Sun distance ($d = 1.47 \times 10^{11}$ m), a frequency of 2.75×10^{12} Hz, an absorption efficiency factor Qabs of 0.7, a dust density   of 3.22 g cm^{-3}, a dust radius a of 1.17×10^{-9} m, and a Planck function $B\lambda(T)$ of 2.28×10^{-15}, we substitute these parameters into the equation to obtain the SiC abundance $M\mathrm{SiC} = 4.75 \times 10^{-5}$, which falls within the corresponding range of 10^{-6}–10^{-3} reported by Ferrarotti & Gail (2006).

4. Conclusions

Through molecular dynamics simulations, we have modeled the coalescence of atoms into small SiC and SiO clusters in AGB stellar winds. We observed that SiC molecules cool from 500 K to 300 K, while SiO molecules cool from 1100 K to 900 K. SiC molecules condense entirely into a single cluster, whereas SiO molecules form multiple small clusters within an evolution time of up to 10^5 fs.

Furthermore, we analyzed the infrared spectra of SiC nanoparticles. The 77SiC, $70Si_3C_3$, and $121Si_3C_3$ models exhibit characteristic peaks at 5, 8.6, 11.3, 15, 19, and 37 m. Specifically, the 77SiC molecule shows peaks at 8.6 and 15 m, the $70Si_3C_3$ molecule shows peaks at 5 and 11.3 m, and the $121Si_3C_3$ molecule shows peaks at 8.6, 19, and 37 m. Many of these peaks correspond to Si–C bond stretching modes.

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